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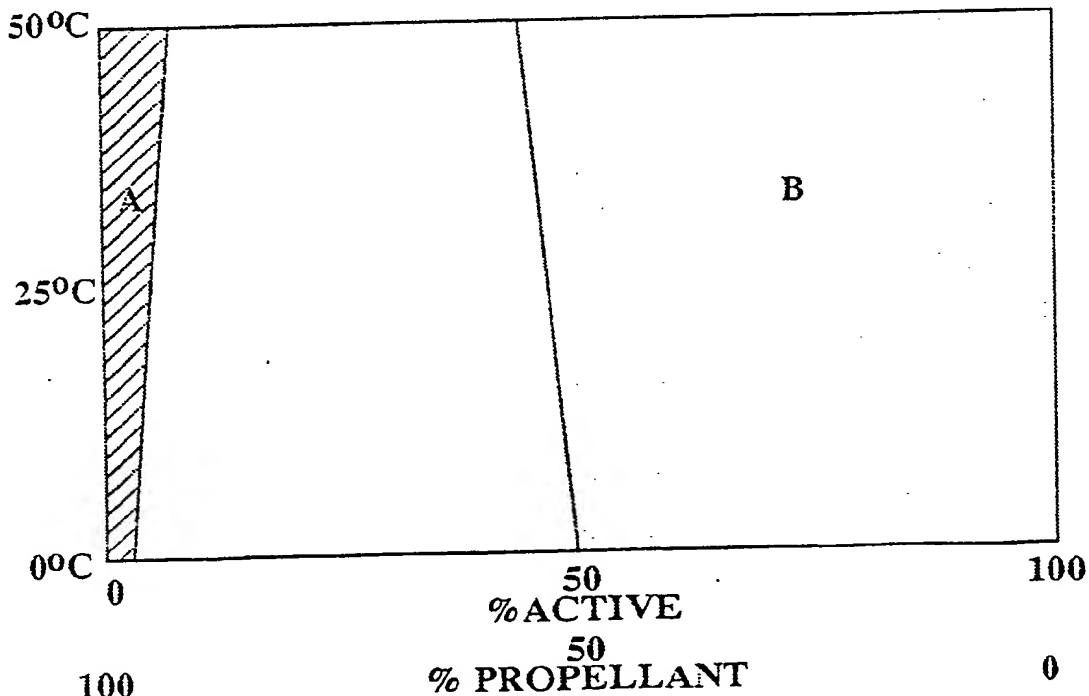
WORLD INTELLECTUAL PROPERTY ORGANIZATION
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INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification ⁵ : C09K 3/30, A01N 25/06 A61K 9/12, A61L 9/14 // B65D 83/54, A01N 43/30, 53/00 A01N 65/00, A61K 7/46	A1	(11) International Publication Number: WO 92/04419 (43) International Publication Date: 19 March 1992 (19.03.92)
(21) International Application Number: PCT/AU91/00396 (22) International Filing Date: 28 August 1991 (28.08.91) (30) Priority data: PK 2089 3 September 1990 (03.09.90) AU (71) Applicant (for all designated States except US): SOLTEC RESEARCH PTY. LTD. [AU/AU]; 8 Macro Court, Rowville, VIC 3178 (AU). (72) Inventor; and (75) Inventor/Applicant (for US only) : TOMLINSON, Roderick, Peter, John [AU/AU]; 32 Highwood Drive, Glen Waverley, VIC 3150 (AU). (74) Agent: WATERMARK; 2nd Floor, The Atrium, 290 Burwood Road, Hawthorn, VIC 3122 (AU).		(81) Designated States: AT (European patent), AU, BE (European patent), CH (European patent), DE (European patent), DK (European patent), ES (European patent), FR (European patent), GB (European patent), GR (European patent), IT (European patent), JP, LU (European patent), NL (European patent), SE (European patent), US. Published <i>With international search report.</i>

(54) Title: A CONCENTRATED AEROSOL SPACE SPRAY



(57) Abstract

A dispenser for space spraying is provided consisting of a container comprising a metering device and a composition essentially comprising 20-85 % w/w active ingredient in adjunct with suitable propellants, solvents and adjuvants. Also provided is an aerosol for use in such a dispenser consisting essentially of 20-85 % w/w active ingredient in adjunct with 80-15 % w/w of suitable propellants, solvents and other adjuvants.

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A CONCENTRATED AEROSOL SPACE SPRAY

The invention relates to an aerosol space spray containing ultra high concentrations of active ingredients.

Throughout this specification the term "space spray" will be used to define aerosol formulations used for dispersion of an active ingredient in the air as opposed to aerosol formulations which are used to apply an active ingredient to a surface, e.g. anti-perspirants, polishes, surface disinfectants etc.

Aerosol formulations have been known for at least 50 years in the art of aerial dispersion of insecticides, air fresheners and other active ingredients.

Conventionally, to obtain the desired characteristics of an aerosol for dispersion into the air or otherwise, the formulations contain minimal active ingredients, a solvent for the active ingredient and a suitable propellant. The active ingredient usually constitutes less than 1% w/w. The solvent is present in the order of 10-20 % w/w and the propellant constitutes 80-90 % w/w. In most cases of such aerosol formulations, the solvent is a hydrocarbon solvent and the propellant a fluorocarbon or hydrocarbon. Alternatively, the propellant is partially substituted with water, wherein the weight percentage being water is in the range of 30-40 % w/w. Therefore it is evident that to deliver one gram of insecticide conventionally requires the release to the atmosphere of between 80 and 250 g of volatile organic compounds (VOC's). Volatile organic compound (VOC) is the general name given for compounds with an appreciable vapour pressure, for example, fluorocarbons, hydrocarbons i.e. kerosene, butane etc. commonly used as propellants.

In surface applications dispersal of the active ingredient of the aerosol over a large area is not vital to the efficacy of the formulation. In many surface sprays it is also important that the active ingredient contact the surface in high concentrations and often for a lengthy period of time - this is particularly important in the case of disinfectants. Accordingly, and in general, aerosol surface sprays containing high amounts of active ingredient are known.

CA 1106329 (BRISTOL-MYERS COMPANY) discloses an antiperspirant formulation comprising about 20-50% w/w of a solid antiperspirant material suspended in about 20-50% w/w of an appropriate oil. About 20-50% w/w of an appropriate propellant is also present in addition to a suspending agent and other adjuvants. In this formulation, the solid active ingredient is suspended in the oil which in turn is suspended throughout the propellant.

Another type of aerosol surface spray is disclosed in JP 78044494 ((TOKA-) TOKYO AEROSOL KAGAKU KAISHA) wherein 65-85 vol % of raw paint liquid, 35-15 vol % of a propellant and a solvent containing not less than 55 vol % (with respect to the paint) of acetone are combined together. The paint is solubilised by the solvent and in turn dissolved in the propellant.

An attempt to increase the amount of powdered solid active ingredient in an aerosol composition was described in AU 512912. In the formulation described at least 15% powdered solid active ingredient is dispersed in an emulsion of water and an anhydrous carrier. Added to this dispersion is then an anhydrous condensed hydrocarbon aerosol propellant gas miscible with the anhydrous carrier so that a final emulsion in which the active ingredient is dispersed is formed. It will be understood that, as in the prior art outlined hereinabove, the active ingredient in this formulation is dispersed or dissolved throughout the propellant. A stated result of this composition is that smaller amounts of propellant gas are required than is conventional for aerosols of this type.

AU 594736 (CHURCH & DWIGHT CO. INC.) discloses an air deodorizer comprising an aerosol having as the dispersed phase, liquid droplets comprising 2-25% of a deodorizing agent. Fragrance may also be included in the deodorizer in an amount of up to 3% by weight of the solution. A propellant in an amount of 8-40% by weight of the resulting aerosol spray may be present as a means for dispersion of the deodoriser.

In a formulation described in AU-B-58959/80 a novel hydrogenated hydrocarbon is described for use as a solvent in aerosol sprays. More particularly 10-89.5% of halogenated hydrocarbon as solvent is claimed in conjunction with 10-70% of liquified aerosol propellant and 0-75% of other organic solvents. Again it is clear that it is the function of the hydrocarbon and other solvents in this formulation to act as solvents of the active ingredient which is present in comparatively small amounts.

According to the prior art, especially in relation to insecticides, in order to solvate pyrethrins and synthetic pyrethroids and other insecticides sufficiently to provide break up energy to fully atomise the insecticide into fine atomised particle dispersions, it is clear that large amounts of propellant and solvents are required in proportion to the active ingredient.

It is apparent that with a general trend around the world for manufacturers and producers to be more environmentally aware as a result of, primarily, Government constraints and consumer demand, there is a real need to reduce the fluorocarbons, hydrocarbons and other VOC's released into the atmosphere since it is thought that they contribute significantly to atmospheric pollution.

Much research has been performed in search of finding a substitute propellant for the conventional fluorocarbon and hydrocarbon propellants for example, by the substitution of water for some of the hydrocarbons; however, a comparative substitute is yet to be found. By way of the water substitute example, water based aerosols do function
5 to disperse the active ingredient but generally not in sufficiently fine enough drops for adequate air dispersal.

Accordingly it is an object of this invention to ameliorate at least some of the disadvantages of the prior art.

More particularly, it is an object of this invention to achieve an improved
10 aerosol space spray which requires less release of undesirable VOC's into the environment. Most desirably, it is another object of the invention to substantially reduce the amount of VOC's required in an aerosol surface spray.

To this end there is provided a dispenser for space spraying consisting of a container comprising a metering device, and a composition essentially comprising 20-
15 85% w/w active ingredient in adjunct with suitable propellants, solvents and adjuvants.

Surprisingly, it has been found that by providing a stable ultra concentrated aerosol, the percentage of undesirable VOC's required as propellants and solvents is dramatically decreased, resulting in less VOC release into the atmosphere. As an added benefit, as a result of the decreased requirement for VOC's, the size of the container used
20 for the aerosol in commercial applications is substantially reduced.

The aerosol arrangement of the invention can be used in essentially any system wherein the active ingredient is to be dispersed into the air or is atomised, for example, insecticides and air fresheners.

In some situations, a solvent may be required in the aerosol, but will generally
25 be in the range of 0-25% w/w.

In the aerosol dispensers according to the invention, the active ingredients preferably act as solvents for the propellant and, as such, the propellant will accordingly be chosen to suit the active ingredients. This invention is partly predicated on the discovery that a small amount of propellant solvated in an insecticide, perfume or
30 other active ingredient achieves effective space dispersion of the active ingredient.

In the case of insecticides, most normal household insecticides are suitable for use in the dispensers of the invention. For example, pyrethrins or synthetic pyrethroids may preferably be used although other conventional insecticides may be envisaged within the scope of the invention.

In the case of air fresheners, again, most conventional perfumes will be suitable for use. The perfume can preferably be used in pure form and as a result thereof an added advantage is lent to such air fresheners inasmuch as the fragrance quality is unadulterated by the presence of high quantities of solvents and emulsifiers present in conventional systems.

Suitable propellants in accordance with the invention include fluorocarbons, hydrocarbons, dimethyl ether and any other propellants regarded as suitable, and in particular, suitable for household aerosols.

Although not essential to the dispensers of the invention, typical solvents which may be used include methylene chloride, III trichloroethane, ethanol, propanol; aromatic compounds such as toluene; glycol ethers such as carbitol; paraffin solvents such as hexane and petroleum solvent blends such as Shellsol T. However, as previously indicated, it is desirable to minimise and where possible obviate the need for these solvents.

In addition to the aforesaid ingredients, other auxiliary agents may be incorporated in the present composition for e.g. anticorrosive agents etc. and in the case of an insecticide, fragrance. Preferably, these adjuvants are present in a quantity of 1-10% w/w. Suitable adjuvants may be perfumes, anti oxidants such as BHT, corrosion inhibitors such as cyclohexylaminenitrate.

Generally, it is envisaged that the aerosol will be used in a domestic situation and thus the aerosol container will meet the standard International pressure requirements.

It is realised that due to the higher concentration of active ingredient, there would possibly be a tendency to spray excess active ingredient into the atmosphere. To circumvent this problem an aerosol metered valve is preferably incorporated with the aerosol of the present invention. A typical dosage will be in the order of 5-500 mg. A more preferred dosage is in the order of 35-155 mg.

Preferably, because of the solvation characteristics of the active ingredients themselves, some specific polymers may be more desirably used as internal gaskets and metering devices in the dispensers of the invention than others. Such suitable polymers are preferably highly solvent resistant. Preferred metering valves for use in the invention may include a Valois DF10-150. Other possible valves may be supplied by COSTER, SEQUIST or EMSON VALVES of the USA.

In another aspect of the invention there is provided a composition for use in a dispenser for space spraying essentially comprising 20-85% w/w active ingredient in adjunct with suitable propellants, solvents and other adjuvants.

Preferably, the composition comprises 20-85% w/w active ingredient and 80-15% w/w propellant. If necessary, an additional solvent in an amount of 0-25% w/w may be present. Other adjuvants and excipients may also be present in an amount of 0-10%.

Suitable active ingredients depending on the purpose of the space spray, propellants, solvents and excipients are outlined hereinabove in relation to the dispensers according to the invention.

In a further embodiment of the invention there is provided a composition for use in a dispenser for space spraying comprising at least one propellant fully solvated in at least one active ingredient.

Preferably, such a composition comprises 80-15% w/w of a propellant which is fully solvated in 20-85% w/w active ingredient.

Compositions according to this aspect of the invention may additionally comprise 0-25% w/w solvent, the solvent also being fully solvated in at least one active ingredient. Conveniently included are also 0-10% w/w of other suitable excipients or adjuvants.

Suitable active ingredients according to the application of the composition in accordance with this embodiment of the invention, suitable propellants, solvents and adjuvants are as described hereinabove in relation to the dispensers according to the invention.

In all aspects of this invention, the active ingredient in the defined range of the invention acts as the more continuous phase wherein the propellant is dispersed or dissolved to form an homogeneous phase.

Since a homogeneous phase is produced it is believed that the propellant is dissolved in the active ingredient.

Figure 1 illustrates a phase diagram for concentrate technology wherein the differences between the different embodiments of this invention and the compositions of the prior art is illustrated.

On the figure A and B are single phase homogeneous solutions.

A is the zone of conventional aerosol formulation with low levels of actives dissolved in high levels of propellants, generally requiring added solvents.

B is the zone defining the new technology whereby the propellant is believed to be dissolved in the active to form homogeneous solutions. The region in between regions A and B is a two phase region unsuitable for use in aerosol formulations.

At 25°C pyrethrin is soluble at less than 1% in hydrocarbon propellants and
5 most perfumes at less than 5%. However as will be shown pyrethrins, synthetic pyrethroids and perfumes form an homogeneous solution across a broad range of concentrations of hydrocarbon propellant.

In the examples given herein below, the propellant solubility at 20°C is measured by observation on a sliding scale wherein "clear" indicates complete
10 solubility of the propellant in the active ingredient, "slight haze" indicates the first visible indications of insolubility whilst "turbid" indicates virtually complete insolubility.

HCP58 hydrocarbon is considered representative of all hydrocarbons available for use as propellants in compositions of the type exemplified hence its choice for the
15 examples given hereinbelow.

In the case where the active ingredient is a perfume, the following examples illustrate the range of formulations possible within the scope of the invention.

In the experimental data given hereinbelow in which turbidity of the composition is measured, samples were packaged in glass aerosols in order that stability could be
20 evaluated.

Example A1

Perfume: 5286 GIVAUDIN

Propellant: HCP58 HYDROCARBON

	<u>% w/w Perfume</u>	<u>% w/w Propellant</u>	<u>Propellant Solubility</u> <u>20°C</u>
	100	0	clear
	90	10	clear
5	80	20	clear
	70	30	clear
	61	39	clear
	60	40	slight haze
	59	41	slight haze
10	58	42	slight haze
	55	45	slight haze
	50	50	haze
	40	60	turbid
	30	70	turbid
15	20	80	turbid
	10	90	turbid
	0	100	clear

It is evident that a concentration in excess of 40% of propellant will result in separation and precipitation of insoluble perfume components. Such insoluble components will probably lead to valve failure in the dispenser and are therefore clearly undesirable.

Given that at least 15% w/w of propellant is required in the compositions according to the invention in order that sufficient expansive energy is provided to the active ingredient, it can be seen that an appropriate composition of this type might include 61-85% w/w perfume and 39-15% w/w propellant.

These weight ranges can be compared with prior art aerosol fragrances wherein levels above approximately 0.5% of fragrances are not used due to the insolubility of the fragrance in the propellant.

Example A2

30 Perfume: NOUVELLE 301
Propellant: HCP58 HYDROCARBON

<u>% w/w Perfume</u>		<u>% w/w Propellant</u>	<u>Propellant Solubility</u> <u>20 °C</u>
100		0	clear
90		10	clear
5	80	20	clear
	70	30	clear
	65	35	clear
	60	40	clear
	59	41	clear
10	58	42	slightly turbid
	55	45	turbid
	50	50	turbid
	40	60	turbid
	30	70	turbid
15	20	80	turbid
	10	90	turbid
	0	100	clear

Again, it is evident that a composition comprised of 58-85% w/w perfume and 42-15% w/w propellant will be homogenous and thus suitable for use.

20 Example A3

In this example the effect of the addition of a solvent upon the compositions according to the invention was tested.

Perfume: NOUVELLE 301

Propellant: HCP58 HYDROCARBON

	<u>% w/w Perfume</u>	<u>% w/w Propellant</u>	<u>% w/w Solvent</u>	<u>Solubility at 20°C</u>
5	20	65	15	slightly turbid
	20	62	18	slightly turbid
	20	61	19	slightly turbid
	20	60	20	clear
	20	55	25	clear
10	40	55	5	slightly turbid
	40	53	7	slightly turbid
	40	52	8	slightly turbid
	40	51	9	clear
	40	50	10	clear

Clearly, the active ingredient, perfume, is an effective substitute for solvents in the compositions of the invention; the higher the level of perfume (40% w/w) the less solvent is required in order to form a single phase homogenous liquid. Conventionally the active ingredient: propellant: solvent ratio is approximately 1:85:15 whereas in this instance the solution is quite clearly an homogenous solution in the ratio of approximately 1:1.25:0.25. It is evident that whereas in the prior art compositions in order to increase the perfume level, the undesirable solvent levels were increased, in fact surprisingly quite the opposite has been found to be true.

In the case where the active ingredient is an insecticide, the following examples illustrate the range of formulations possible within the scope of the invention.

These results can be compared to a prior art household insecticide formulation which conventionally comprises:

25	Pyrethrins (active)	1.5% w/w
	Piperonyl Butoxide	
	(pyrethroid synergist)	1.5% w/w
	ShellSolT (solvent)	15.0% w/w
	Trichloroethane (solvent)	25.2% w/w
30	HLP58 Hydrocarbon	
	(propellant)	58.0% w/w

i.e. the active ingredient: propellant: solvent ratio is approximately 1:22:32.

In each of the examples given hereinbelow the formulations were prepared in glass aerosols and their clarity examined after 24 hours.

Example B1

Insecticide: 50% KENYA PYRETHRUM EXTRACT

5 Synergist: 85% PIPERONYL BUTOXIDE

(The insecticide and the synergist were combined in a ratio of 1:4)

Propellant: HCP58 HYDROCARBON

	<u>% w/w Insecticide</u>	<u>% w/w Propellant</u>	<u>Solubility 20°C</u>
	100	0	clear
10	90	10	clear
	80	20	clear
	70	30	clear
	60	40	clear
	50	50	clear
15	40	60	clear
	39	61	slight haze
	38	62	slight haze
	37	63	slight haze
	35	65	slight haze
20	30	70	slight haze
	20	80	turbid
	10	90	turbid
	0	100	clear

25 Again, on the understanding that at least 15% w/w propellant is required in order that sufficient expansion energy is provided to the active ingredient, it can be seen that a composition suitable for use can be comprised of 40-85% w/w insecticide and 60-15% w/w propellant.

Example B2

30 Insecticide: SUMETHRIN

Propellant: HCP58 HYDROCARBON

	<u>% w/w Insecticide</u>	<u>% w/w Propellant</u>	<u>Solubility 20°C</u>
	100	0	clear
	90	10	clear
	80	20	clear
5	79	21	slight haze
	78	22	slight haze
	76	24	smoky
	70	30	smoky
	60	40	smoky
10	50	50	smoky
	40	60	turbid
	30	70	turbid
	20	80	turbid
	10	90	turbid
15	0	100	clear

It can be seen that suitable compositions might comprise 80-85% w/w insecticide and 20-15% w/w propellant.

In order to test the effect of the presence of a solvent on this composition, 5% w/w solvent was added to the composition. This amount of solvent enabled a solution of 20 50% w/w insecticide and 45% w/w propellant which was completely clear and thus suitable for use.

Example B3

Insecticide: BIORESMETHRIN/BIOALLETHRIN 1:5

Propellant: HCP HYDROCARBON 58

	<u>% w/w Insecticide</u>	<u>% w/w Propellant</u>	<u>Solubility 20°C</u>
	100	0	clear
	90	10	clear
	80	20	clear
5	70	30	clear
	68	32	clear
	66	34	clear
	64	36	clear
	63	37	slightly turbid
10	62	38	slightly turbid
	60	40	turbid
	50	50	turbid
	40	60	turbid
	30	70	turbid
15	20	80	turbid
	10	90	turbid
	0	100	clear

Satisfactory atomisation of the insecticide is achieved in compositions having 64-85% w/w insecticide and 36-15% w/w propellant.

- 20 Addition of 5% w/w solvent enabled a higher quantity of propellant to be dissolved in the insecticide.

Example C1

- In order to test the efficacy of the compositions according to the invention, the composition described in Example B1 in a ratio of 50% w/w insecticide blend to 50% w/w propellant was packaged in a 10 mg aluminium aerosol and fitted with a 150 mg metering valve.

Products

1. Formulation - Aerosol Insecticide of Example B1

Methods

- 30 For these tests, 5 replicates only were carried out for the Example B1 formulation using the CERIT Modified Hunting Mode protocol - 'CERIT/HF-HM/FIK 2.0' - modified for manual operation and to allow the aerosol plume of the formulation to envelop the fly release cage fully.

MODIFIED HUNTING MODE PROTOCOL -
AEROSOL CHAMBER TESTS-CERIT/HF-HM/FIK 2.0

Hunting Mode

Insects released to fly through the spray cloud, to simulate field use where the operator "aims at" the target(s), i.e. the "Hunting" method.

Order of Testing

The order of testing of formulations was chosen on a randomized block basis. Each formulation, including controls, was tested in each block; each block being completed on one day.

10 Five replicates of each test.

Controls

One run in each replicate block, without aerosol spray.

Insects

Houseflies - *Musca domestica*

15 Strain - "SYD 90", field collected 1990, from Sydney, various areas.

Resistance status - susceptible (equivalent to SYD 88).

Pre-fed sugar only, no protein.

Age at time of test - 3 to 7 days.

Sex - mixed.

20 Transferred directly from cage to release container without anaesthesia.

Numbers used per test - approx. 50.

Sprays

Formulation - supplied as a very small aerosol.

25 Spray rate - pre-calibration of aerosol before use; aerosol shaken (inverted) immediately before spraying.

Spray start - at time zero.

Spray duration - manually.

Weight sprayed - 0.08 g.

Insect Release

30 Released into chamber - mechanically, under computer control, 0.8 m in front of , and 20 cm above, nozzle.

Time of release - 2s.

K.D. Counts.

Counting - visually.

Times of counts (from time zero). in seconds:

30, 60, 90, 120, 150, 180, 240, 360, 480, 720.

5 Evacuation of Aerosol

Vents opened and exhaust fan on for 15 mins after each test.

Recording

Computer, disk and printout.

Preliminary Analyses

- 10 Log dose/probit analysis - a modified program.
KDT₅₀ s and 95% confidence limits are determined.
Data then subjected to analysis of variance program and Student-Newman
Keuls test.

Mortality

- 15 Insects held in containers provided with sugar and water for 24 h counts of
mortality.

The modifications to this protocol for the purposes of this example were as
follows:

1. a reduction in spraying distance from 1.8m to 0.8m because of the low plume
20 projection or 'throw' of the formulation;
2. a reduction in the standard 2 g delivery of insecticide to a metered dose delivery
of approximately 0.08 g;
3. and manual actuation of the formulation instead of computer-controlled actuation.

Analyses

- 25 The times for 50% of the insects to be knocked down (KDT₅₀ s) were calculated
by probit analysis (Finney, D.J., 1971. Probit Analysis. 3rd ed. Cambridge Univ. Press,
London. 333pp). Analysis of variance (ANOVA - Sokal and Rohlf "Biometry", Freeman,
1981) was applied to the KDT₅₀ s for all formulations.

Results

TABLE 1. KDT₅₀ s (sec) and the 24 h mortalities (%) for Modified Hunting Mode, 1 metered dose per run = 0.08 g spray, 2 sec release.

5 Formulation	Mean KDT ₅₀	95% C-Limits	Mean Mortality
	(sec)	(sec)	(%)
Supersol	116	108-124	100

In the tests for Modified Hunting Mode, the formulation of Example B1 was effective in the knockdown and kill of *Musca domestica*, where a KDT₅₀ of 116 seconds and 100% mortality was achieved (from Table 1).

10 Based on the efficacies of formulations that are available on the market, the results for the formulation of Example B1 are comparable with mid-range products, i.e. total knockdown and mortality (100%) is achieved within 2 minutes of spraying.

It is apparent from the examples given hereinabove that whereas in conventional household aerosols, solvent/insecticide ratios can be as high as 100:1 and propellant 15 ratios even higher, in the formulations of the instant invention, the solvent/insecticide ratio is less than 1:5 and often zero and the propellant ratios less than 1:1.

It is also therefore apparent that in accordance with these embodiments wherein the active ingredient is either an insecticide or a perfume, the delivery of one gram of active component is accompanied by the release of only one gram of VOC's, or indeed less. 20 This represents a 100 fold reduction in VOC release in comparison to prior art formulations.

It is to be understood that the preceding examples are intended to illustrate but not limit the compositions and dispensers of the instant invention. It will be understood that the scope of the invention may include such active ingredients as anti-microbial 25 agents and may be extended to include other solvents, adjuvants and propellants not specifically illustrated.

THE CLAIMS DEFINING THE INVENTION ARE AS FOLLOWS:

1. A dispenser for space spraying consisting of a container comprising a metering device, and a composition essentially comprising 20-85% w/w active ingredient in adjunct with suitable propellants, solvents and other adjuvants.
2. A dispenser as claimed in claim 1 wherein said composition comprises 20-85% w/w active ingredient and 80-15% w/w propellant.
3. A dispenser as claimed in claims 1 or 2 wherein said composition comprises 0-25% w/w solvent.
4. A dispenser as claimed in any one of claims 1, 2 or 3 wherein said composition additionally comprises 0-10% w/w of other suitable adjuvants.
5. A dispenser as claimed in any one of claims 1-4 wherein said propellant is selected from the group consisting of hydrocarbons, fluorocarbons and dimethyl ether.
6. A dispenser as claimed in any one of claims 1-5 wherein said active ingredient consists of an insecticide and/or a perfume.
7. An aerosol for use in a dispenser for space spraying consisting essentially of 20-85% w/w active ingredient in adjunct with 80-15% w/w of suitable propellants, solvents and other adjuvants.
8. A composition as claimed in claims 7 comprising 0-25% w/w solvent.
9. A composition as claimed in any one of claims 7 or 8 comprising 0-10% w/w of other suitable adjuvants.
10. A composition as claimed in any one of claims 7-10 wherein said active ingredient consists of an insecticide and/or a perfume.
11. A composition as claimed in any one of claims 7-10 wherein said propellant is selected from the group comprising hydrocarbons, fluorocarbons and dimethyl ether.

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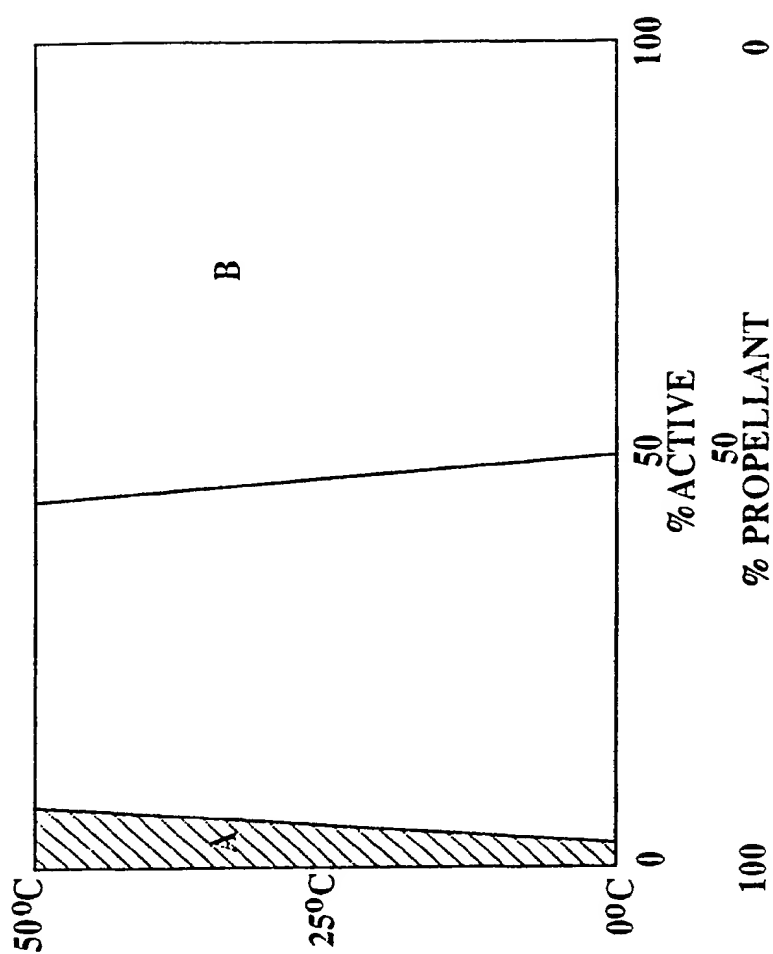


Fig 1.

INTERNATIONAL SEARCH REPORT

I. CLASSIFICATION OF SUBJECT MATTER (if several classification symbols apply, indicate all) ⁶												
According to International Patent classification (IPC) or to both National Classification and IPC Int. Cl. ⁵ C09K 3/30, A01N 25/06, A61K 9/12, A61L 9/14 // B65D 83/54, A01N 43/30, 53/00, 65/00, A61K 7/46												
II. FIELDS SEARCHED												
Minimum Documentation Searched ⁷												
Classification System	Classification Symbols											
Int. Cl. ⁵	C09K 3/30, A01N 25/06, A61K 9/12											
Documentation Searched other than Minimum Documentation to the extent that such Documents are included in the Fields Searched ⁸												
AU: Int. Cl. as above												
III. DOCUMENTS CONSIDERED TO BE RELEVANT ⁹												
Category [*]	Citation of Document, ¹¹ with indication, where appropriate of the relevant passages ¹²	Relevant to Claim No ¹³										
X	US, A, 4851212 (WINSTON et al.) 25 July 1989 (25.07.89) See abstract.	1-11										
X	Derwent Soviet Inventions Illustrated, Section I, Chemical, Issued October 1965, General Organic, p. 2, SU 166861 (FEDDER et al.) August 1965 (00.08.65) See abstract.	1-11										
X	Patent Abstracts of Japan, C-235, page 117, JP,A,59-64688 (JIYONSON K.K.) 12 April 1984 (12.04.84) See abstract.	1-11										
(continued)												
<p>[*] Special categories of cited documents : ¹⁰</p> <table border="0"> <tr> <td>"A" Document defining the general state of the art which is not considered to be of particular relevance</td> <td>"T" Later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention</td> </tr> <tr> <td>"E" earlier document but published on or after the international filing date</td> <td>"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step</td> </tr> <tr> <td>"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)</td> <td>"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art</td> </tr> <tr> <td>"O" document referring to an oral disclosure, use, exhibition or other means</td> <td>"&" document member of the same patent family</td> </tr> <tr> <td>"P" document published prior to the international filing date but later than the priority date claimed</td> <td></td> </tr> </table>			"A" Document defining the general state of the art which is not considered to be of particular relevance	"T" Later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention	"E" earlier document but published on or after the international filing date	"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step	"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)	"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art	"O" document referring to an oral disclosure, use, exhibition or other means	"&" document member of the same patent family	"P" document published prior to the international filing date but later than the priority date claimed	
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IV. CERTIFICATION												
Date of the Actual Completion of the International Search 8 November 1991 (08.11.91)	Date of Mailing of this International Search Report 13 November 91											
International Searching Authority AUSTRALIAN PATENT OFFICE	Signature of Authorized Officer SIAW-JAN YONG											

III. DOCUMENTS CONSIDERED TO BE RELEVANT (CONTINUED FROM THE SECOND SHEET)

Category*	Citation of Document, ¹¹ with indication, where appropriate of the relevant passages ¹²	Relevant to Claim No ¹³
X	Patent Abstracts of Japan, C-537, page 131, JP,A, 63-137981 (KANEBO LTD) 9 June 1988 (09.06.88) See abstract.	1-11
X	Patent Abstracts of Japan, C-707, page 65, JP,A, 2-24382 (OSAKA AEROSOL IND CORP) 26 January 1990 (26.01.90) See abstract.	1-11
X	Patent Abstracts of Japan, C-723, page 112, JP,A, 2-69407 (TAIYO KAGAKU CO LTD) 8 March 1990 (08.03.90) See abstract.	1-11

ANNEX TO THE INTERNATIONAL SEARCH REPORT ON
INTERNATIONAL APPLICATION NO. PCT/AU 91/00396

This Annex lists the known "A" publication level patent family members relating to the patent documents cited in the above-mentioned international search report. The Australian Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

Patent Document Cited in Search Report		Patent Family Member			
US	4851212	AU 67673/87	CA 1279577	EP	231084
		NZ 218183	PH 23849	US	4740366

END OF ANNEX